Optical/Dielectric Properties of Inhomogeneous Materials: A New Method of Evaluation

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ABSTRACT

A new method, based on Effective Mean Field Theory, is proposed for the evaluation of the optical/dielectric properties of inhomogeneous materials, in which the real and imaginary parts of the dielectric function are determined by solving a simultaneous non-linear equation. It is seen that the method yields precise and unambiguous results and can be applied to any type of inhomogeneous material.

Key words: optical, Dielectric, Inhomogeneous, Effective Mean Field Theory, Composite Films.

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1. Introduction:

The optical/dielectric properties of materials can be modified by mixing two or more materials in different proportions.[1-3] The constituent materials can be metals, semiconductors and insulators which may be either in solid[1,2] or liquid[3] form. One of the effective ways of producing inhomogeneous materials in the solid state is by co-evaporation or sputtering of the constituent materials which has been widely investigated.[4-6] These materials have useful applications, as solar absorbers.[7] The optical/dielectric properties of liquids can be altered by mixing different proportions of metallic particles in the liquid.[8] Such liquid-solid inhomogeneous systems have been studied by Cohen and his workers.[8,9]

Irrespective of the system under study, one of the important aspects of these investigations is to theoretically evaluate the optical and dielectric constants from the knowledge of the optical/dielectric constants of the constituent materials for which two well known theories, namely, the Maxwell-Garnet theory[10] and the Effective Mean Field Theory (EMT) due to Bruggeman[11] are available. The physical/mathematical basis of these two theories and the topological conditions under which these two theories are applicable have been discussed by various investigators.[12-14]

EMT due to Bruggeman, which was originally developed considering topologically symmetric morphology[11,12] shows better agreement with experiments in many of the solid-solid binary inhomogeneous materials.[13,14] It is also equally good in predicting the dielectric functions of binary liquid-solid composite materials.[8,9] While a definite method exists for evaluation of the average optical/dielectric constants of the inhomogeneous materials from the knowledge of the optical/dielectric properties of the constituent materials, there are certain drawbacks such as limited accuracy and applicability. The objective of the present paper is to arrive at a unique and novel evaluation scheme which is not only more accurate; but also more amenable to the form of existing problems.

2. Theory

The dielectric constant of a multi-component inhomogeneous material according to EMT due to Bruggeman[11,12] is given by:

$$\sum_{i=1}^{n} \frac{3F_i}{2 + \epsilon_i/\langle\epsilon\rangle} = 1 \tag{1}$$

where $\langle\epsilon\rangle$ is the average dielectric constant of the inhomogeneous material

ϵ_i is the dielectric constant of the i^{th} component material

F_i is the volume fraction of the i^{th} component material

and

$$\sum_{i=1}^{n} F_i = 1 \tag{2}$$

Let ϵ_i be given by $\epsilon_i = \epsilon_{iR} + j\epsilon_{iI}$ where ϵ_{iR} and ϵ_{iI} are the real and imaginary parts of ϵ_i, respectively. Then Eqn. (1) can be re-written as

$$\sum_{i=1}^{n} \frac{3F_i}{2 + (\epsilon_{iR} + j\epsilon_{iI})/\langle\epsilon\rangle} = 1 \tag{3}$$

and expanding:

$$3F_1 \prod_{i=2}^{n} \left(2 + (\epsilon_{iR} + j\epsilon_{iI})/\langle\epsilon\rangle \right) + 3F_2 \prod_{\substack{i=1 \\ (i\neq 2)}}^{n} \left(2 + (\epsilon_{iR} + j\epsilon_{iI})/\langle\epsilon\rangle \right) + \ldots$$

$$+ 3F_n \prod_{i=1}^{n-1} \left(2 + (\epsilon_{iR} + j\epsilon_{iI})/\langle\epsilon\rangle \right) = \prod_{i=1}^{n} \left(2 + (\epsilon_{iR} + j\epsilon_{iI})/\langle\epsilon\rangle \right) \tag{4}$$

It is proposed that the real and imaginary parts of Eqn. (4) be separated and the resulting simultaneous non-linear equation which consists of the two unknowns, namely $\langle\epsilon_R\rangle$ and $\langle\epsilon_I\rangle$, and known ϵ_{iR} and ϵ_{iI}, is solved to arrive at $\langle\epsilon_R\rangle$ and $\langle\epsilon_I\rangle$.

Eqn. (4) for a binary composite system (n=2) turns out to be

$$3F \left(2 + (\epsilon_{1R} + j\epsilon_{1I})/\langle\epsilon\rangle \right) + 3(1-F) \left(2 + (\epsilon_{2R} + j\epsilon_{2I})/\langle\epsilon\rangle \right)$$

$$= \left(2 + (\epsilon_{1R} + j\epsilon_{1I})/\langle\epsilon\rangle \right) \left(2 + (\epsilon_{2R} + j\epsilon_{2I})/\langle\epsilon\rangle \right) \tag{5}$$

In Eqn. (5), F is the volume fraction of the material whose dielectric constant is ϵ_2. After a few algebraic manipulations, Eqn. (5) can be simplified and written as

$$\langle\epsilon\rangle + A = B/\langle\epsilon\rangle \tag{6}$$

where $A = 1/2\ [\epsilon_{1R}(3F-2) + \epsilon_{2R}(1-3F) + j\{\epsilon_{1I}(3F-2) + \epsilon_{2I}(1-3F)\}]$

and $B = 1/2\ [\{\epsilon_{1R}\epsilon_{2R} - \epsilon_{1I}\epsilon_{2I}\} + j\{\epsilon_{1I}\epsilon_{2R} + \epsilon_{2I}\epsilon_{1R}\}]$

In the earlier approaches, equation (6) which is quadratic in

$\langle\epsilon\rangle$ is solved,[8,9,13,15] and then the real and imaginary parts of $\langle\epsilon\rangle$ are separated out. On the other hand, in the present approach, the real and imaginary parts of equation (6) are separated out first. Substituting $A_R + jA_I$ for A, $B_R + jB_I$ for B, and $\langle\epsilon_R\rangle + j\langle\epsilon_I\rangle$ for $\langle\epsilon\rangle$ in Eqn. (6) and multiplying and dividing by the complex conjugate of $\langle\epsilon\rangle$, it is possible to write the following two equations:

$$\langle\epsilon_R\rangle^3 + \langle\epsilon_R\rangle^2 A_R + \langle\epsilon_I\rangle^2\langle\epsilon_R\rangle + A_R\langle\epsilon_I\rangle^2 - B_R\langle\epsilon_R\rangle - B_I\langle\epsilon_I\rangle = 0 \quad (7)$$

$$\langle\epsilon_I\rangle^3 + \langle\epsilon_I\rangle^2 A_I + \langle\epsilon_R\rangle^2\langle\epsilon_I\rangle + A_I\langle\epsilon_R\rangle^2 - B_I\langle\epsilon_R\rangle + B_R\langle\epsilon_I\rangle = 0 \quad (8)$$

Eqns. (7) and (8) are non-linear algebraic equations in $\langle\epsilon_R\rangle$ and $\langle\epsilon_I\rangle$ and the solutions can be determined by a numerical iterative technique.[16] It is proposed to adapt the Newton-Raphson iterative method [16,17] and the corresponding iteration formulae are given as follows:

$$\langle\epsilon_R(i+1)\rangle = \langle\epsilon_R(i)\rangle + \frac{f_2\dfrac{\partial f_1}{\partial\epsilon_I} - f_1\dfrac{\partial f_2}{\partial\epsilon_I}}{\dfrac{\partial f_1}{\partial\epsilon_R}\dfrac{\partial f_2}{\partial\epsilon_I} - \dfrac{\partial f_2}{\partial\epsilon_R}\dfrac{\partial f_1}{\partial\epsilon_I}} \quad (9)$$

$$\langle\epsilon_I(i+1)\rangle = \langle\epsilon_I(i)\rangle + \frac{f_1\dfrac{\partial f_2}{\partial\epsilon_R} - f_2\dfrac{\partial f_1}{\partial\epsilon_R}}{\dfrac{\partial f_1}{\partial\epsilon_R}\dfrac{\partial f_2}{\partial\epsilon_I} - \dfrac{\partial f_2}{\partial\epsilon_R}\dfrac{\partial f_1}{\partial\epsilon_I}} \quad (10)$$

where $\langle\epsilon_R(i)\rangle$, $\langle\epsilon_I(i)\rangle$ and $\langle\epsilon_R(i+1)\rangle$, $\langle\epsilon_I(i+1)\rangle$ are the respective real and imaginary parts of the dielectric constant $\langle\epsilon\rangle$ for the i^{th} and $(i+1)^{th}$ iterations; and f_1 and f_2 are respectively given by

$$f_1 = \langle\epsilon_R\rangle^3 + \langle\epsilon_R\rangle^2 A_R + \langle\epsilon_I\rangle^2\langle\epsilon_R\rangle + A_R\langle\epsilon_I\rangle^2 - B_R\langle\epsilon_R\rangle - B_I\langle\epsilon_I\rangle$$

and $$f_2 = \langle\epsilon_I\rangle^3 + \langle\epsilon_I\rangle^2 A_I + \langle\epsilon_R\rangle^2\langle\epsilon_I\rangle + A_I\langle\epsilon_R\rangle^2 - B_I\langle\epsilon_R\rangle + B_R\langle\epsilon_I\rangle$$

A closed loop iteration defined by eqns. (9) and (10) for 10 to 15 cycles results in f_1 and f_2 converging to zero and solutions for $<\epsilon_R>$ and $<\epsilon_I>$ are arrived at unambiguously.

As can be seen, $<\epsilon_R>$ and $<\epsilon_I>$ are evaluated in an unambiguous way. In the earlier approaches,[8,9,13,15] a quadratic solution of equation (6) is arrived at resulting in an ambiguity of dual solutions, which is, however, avoided by the stipulated condition that $<\epsilon_I>$ be always real (i.e. $<\epsilon_I> = \mathrm{Img}(<\epsilon>) \geq 0$). In so doing, the argument under the square root is treated as "completely real".[8,9]

3. Results and Discussion:

The procedure is applied to evaluate $<\epsilon_R>$ and $<\epsilon_I>$ for a dielectric--metal binary composite system in which the dielectric constants of the individual components are given by $\epsilon_1 = 1.0 + j0.0$ and $\epsilon_2 = 1.0 - \omega_p^2/\omega(\omega + j\gamma)$ where ω_p is the plasma frequency and γ is the inverse relaxation time.[9] γ/ω_p is set equal to 0.4. The calculations using our method are carried out for different volume fractions and the results are shown in Figs. 1a - 1c and compared with those due to Webman, et. al.[9] From the figure it is evident that the two sets of results agree with each other at lower volume fractions (F = 0.046), whereas at higher volume fractions (F = 0.15 and 0.25) they differ. The difference is maximum in regions close to the resonance frequency and is as high as 15% and 8% for $<\epsilon_R>$ and $<\epsilon_I>$ respectively (with F = 0.25).

To further examine the accuracy of the evaluation techniques, both methods are applied to evaluate the dielectric function of an inhomogeneous system consisting of metallic and non-metallic materials whose dielectric properties are given by[8]

$$\epsilon_1 = 1 - f_i/[(\omega^2 - \omega_0^2) + i\omega\gamma] \text{ and } \epsilon_2 = 1 - \omega_p^2/\omega(\omega + i/\tau).$$

The metallic regions are characterized by the plasma frequency and. relaxation time τ with $\omega_p\tau = 4.0$. While the non-metallic regions are specified by the oscillator strength $f_i\omega_p^{-2} = 0.6$, the resonance frequency $\omega_0/\omega_p = 0.6$ and the resonance width by $\tau/\omega_p = 0.25$.[8] As is evident from the results of the calculations presented in Figs. 2a-2c, there are differences between the two sets of results. The magnitude of the difference is as high as 45% in $\langle\epsilon_I\rangle$ and 6% in $\langle\epsilon_R\rangle$ for the case of F = 0.8; and 15% in ϵ_I and 35% in ϵ_R for F = 0.5. For F = 0.2, they are respectively equal to 35% and 23%.

The results between the present and earlier evaluation techniques differ significantly at higher volume fractions . The difference is greater for a composite system in which both components have imaginary contributions to the dielectric constant, as exhibited in Fig. 2. In the present technique, the contributions to the average dielectric properties, from the components of the composite material, are appropriately accounted for, by introducing the relevant terms in the constituent equation. On the other hand, in the earlier approach[8,9] there may be a possibility that contributions from the imaginary part of the dielectric constants are not properly accounted for thereby contributing to the errors in the final calculations. In view of these observations, the present method has scope for the precise evaluation of inhomogeneous materials. In addition, the present technique is easily extended to systems consisting of more than two components by modifying the constituent equations. It has been applied to study the optical properties of inhomogeneous thin films of Ge:Ag and Si:Au in the infrared region, the results of which will be presented elsewhere.[18]

4. Conclusions:

A new method for evaluation of the dielectric properties of inhomogeneous materials has been successfully developed. The method has scope for precise and unambiguous evaluation and can be applied '/ extended to any type of inhomogeneous material which can be mainly attributed to the unique scheme of evaluation adapted.

The work described in this paper was performed by the Center for Space Microelectronics Technology, Jet Propulsion Laboratory, California Institute, of Technology, under sponsorship of the National Aeronautics and Space Administration, Office of Advanced Concepts and Technology (NASA/OACT). One of the authors (CLN) would like to thank the Department of Science and Technology and the Indian Space Research Organization of the government of India and the United States Agency for International Development (USAID), government of India, for the necessary financial support and encouragement. Also, he is thankful to the Academy for Educational Development (AED), U.S.A. for logistic support and encouragement.

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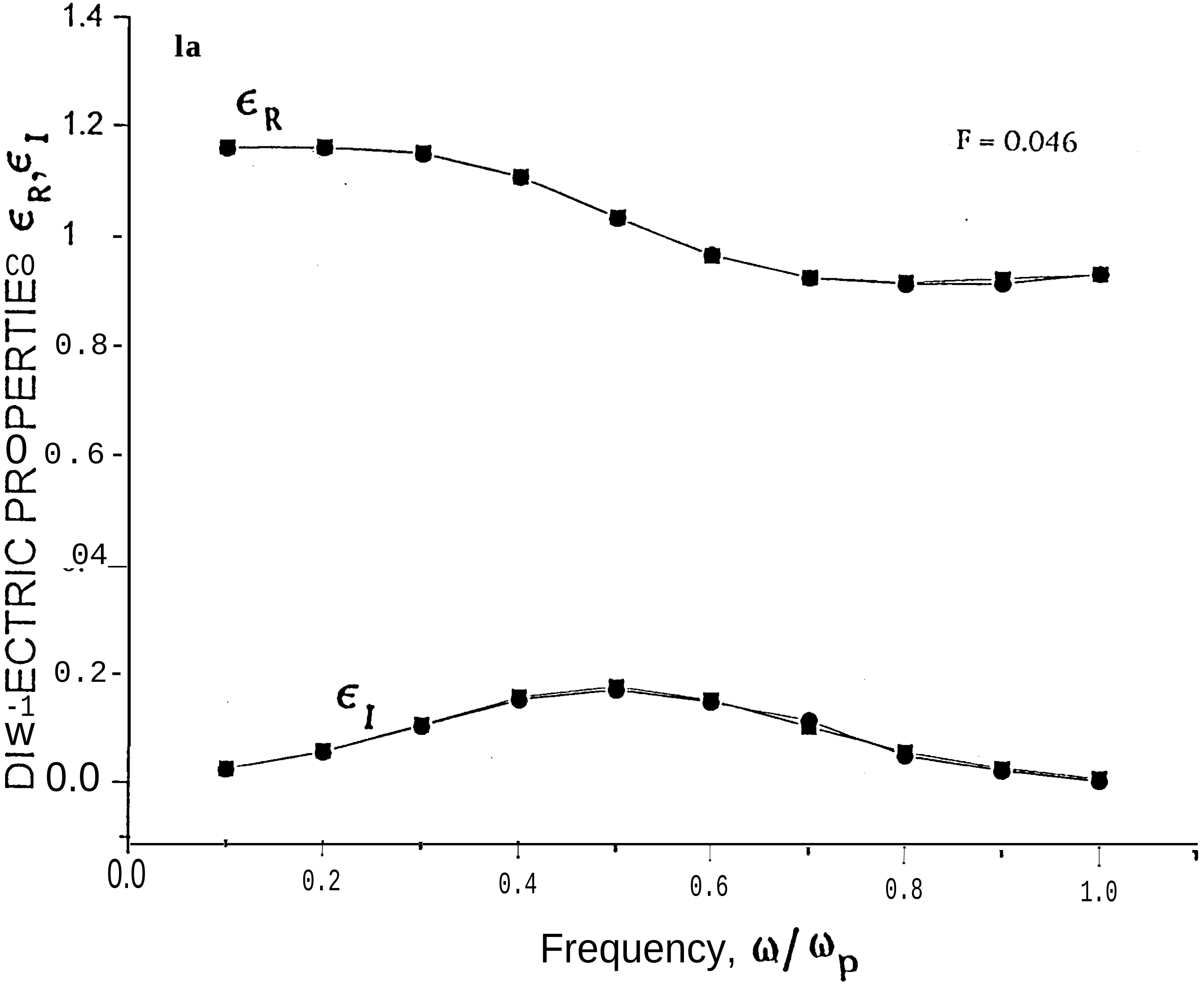
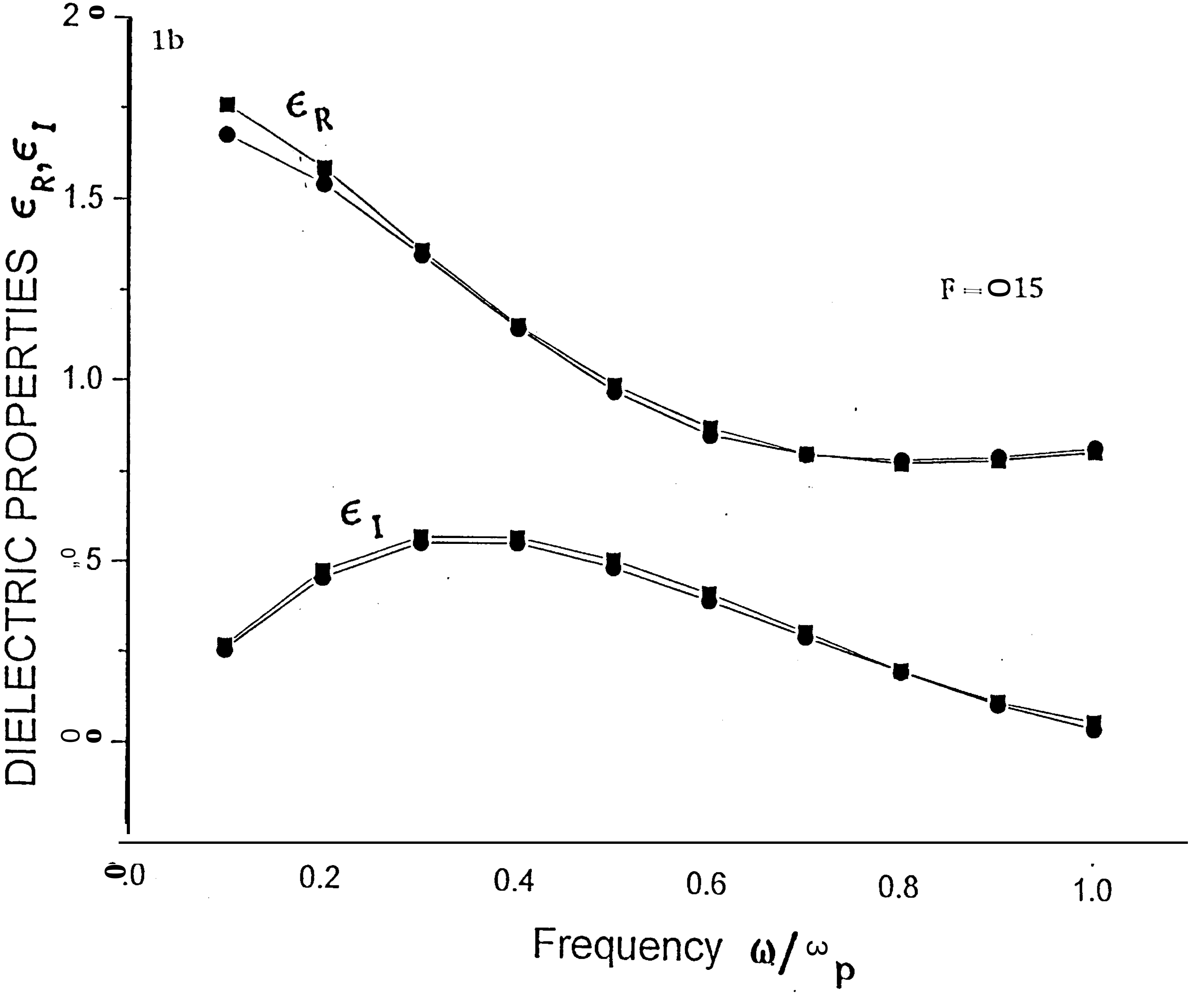
Figure Captions:

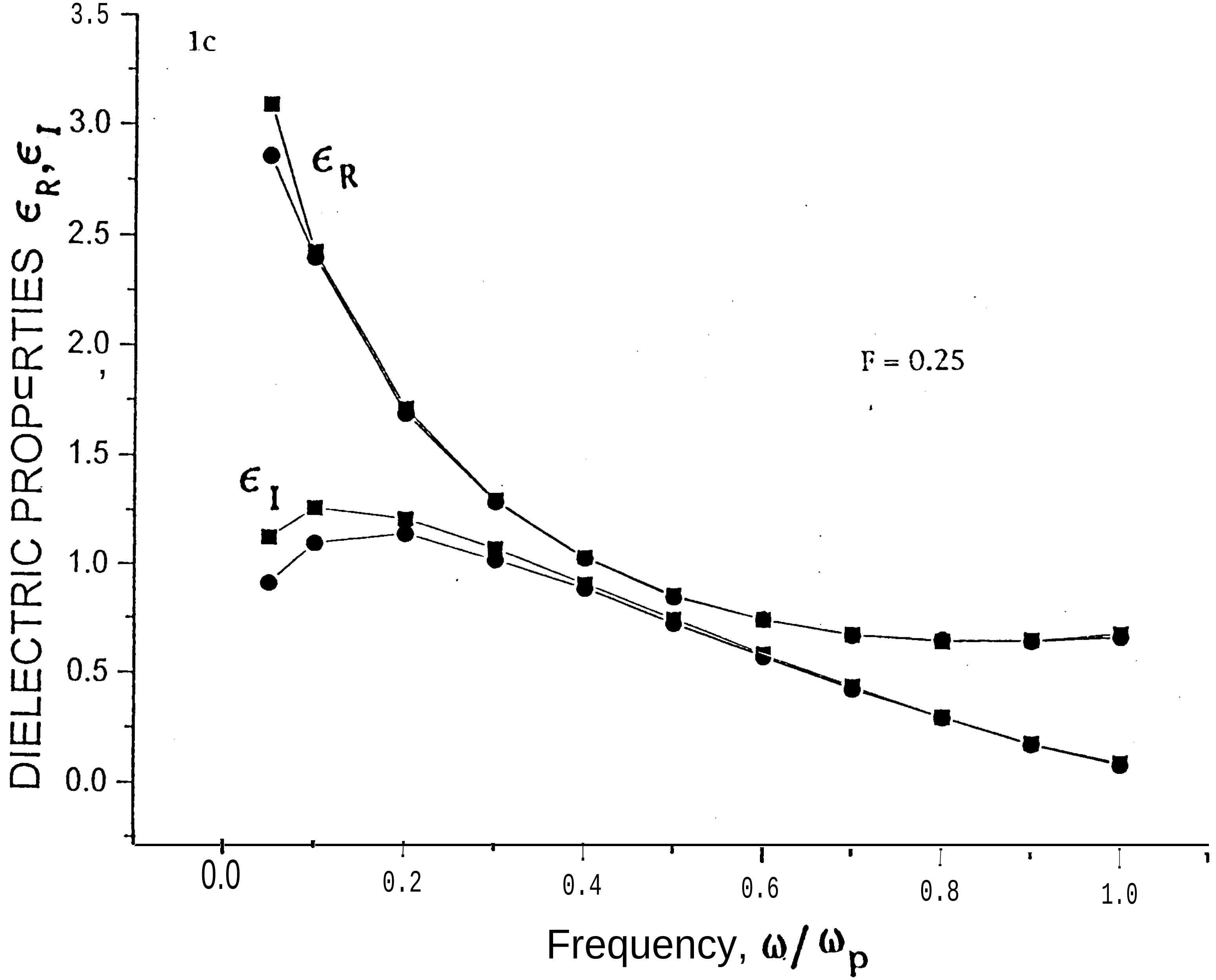
Figs. 1a - 1c: Real and imaginary parts of the dielectric function of a dielectric-metal binary composite material. ■ Present method. ● From Ref. 9.

Figs. 2a - 2c: Real and imaginary parts of a two component composite system." The dielectric function of both components are imaginary. ■ Present method. ● From Ref. 8.

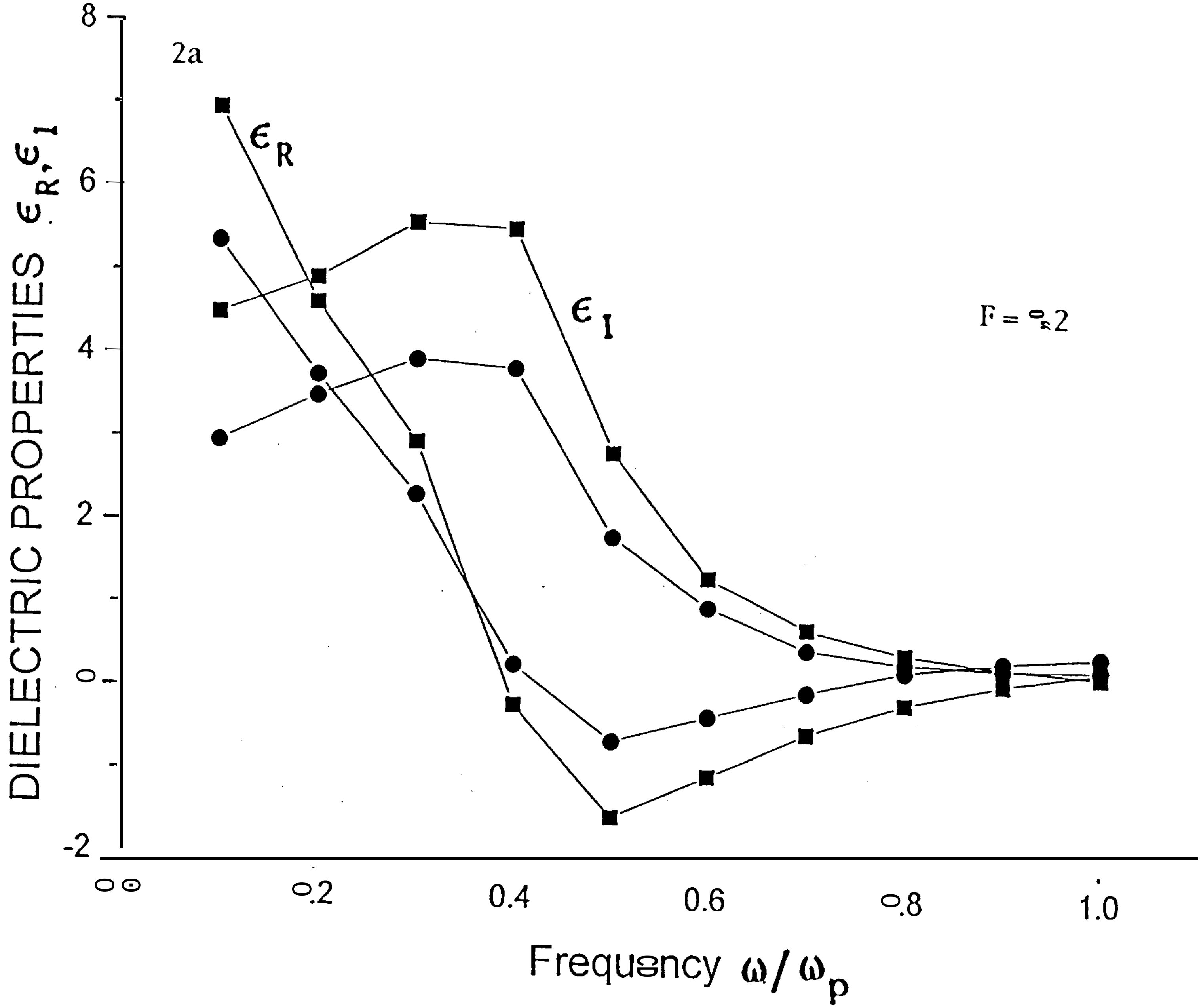


la
ϵ_R
F = 0.046
ϵ_I
DIW-1ECTRIC PROPERTIECO ϵ_R, ϵ_I
1.4
1.2
1
0.8
0.6
04
0.2
0.0
0.0
0.2
0.4
0.6
0.8
1.0
Frequency, ω/ω_p

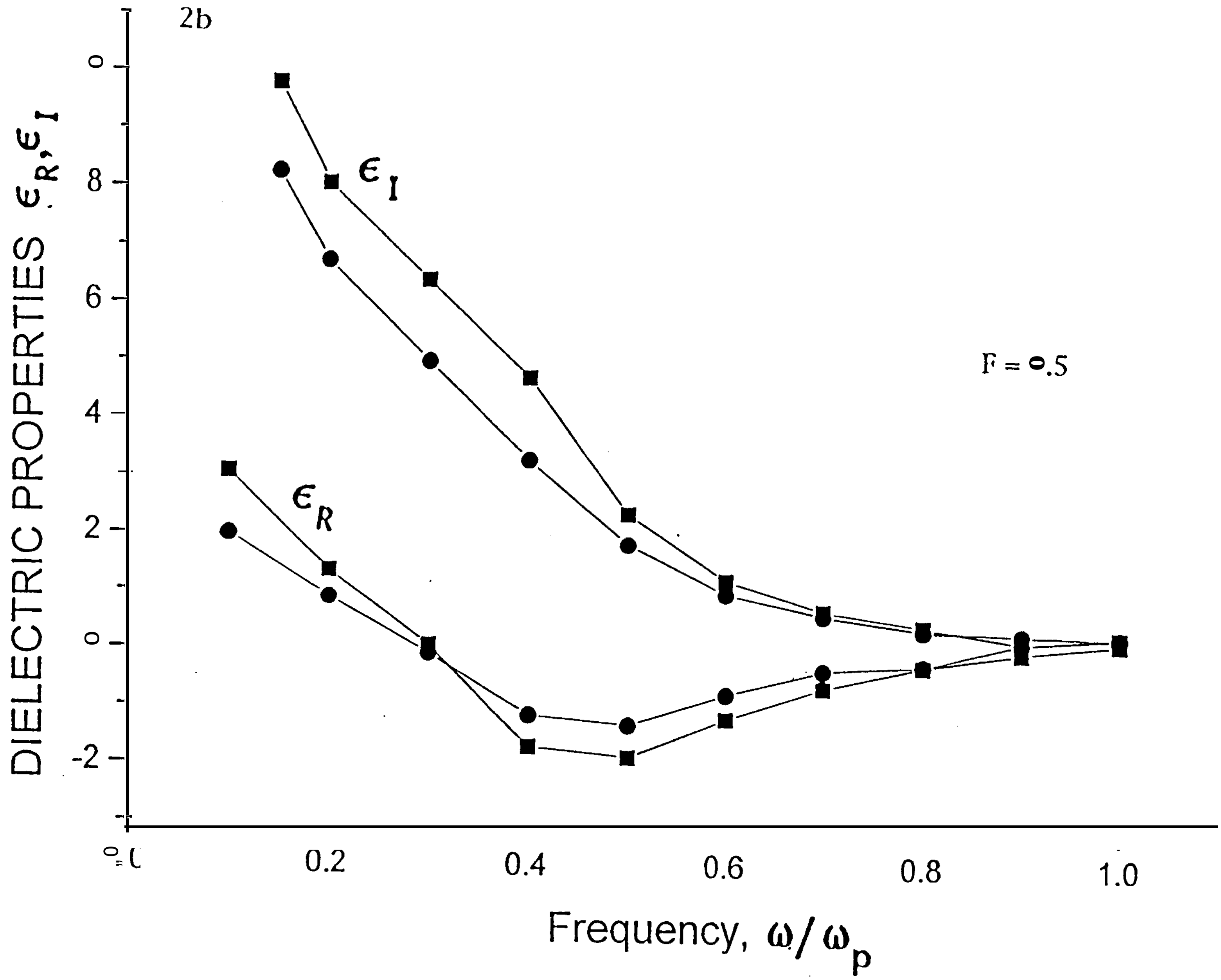

1b
F = O15
ϵ_R
ϵ_I
DIELECTRIC PROPERTIES ϵ_R, ϵ_I
Frequency ω/ω_p
2.0
1.5
1.0
.5
0
0.0
0.2
0.4
0.6
0.8
1.0

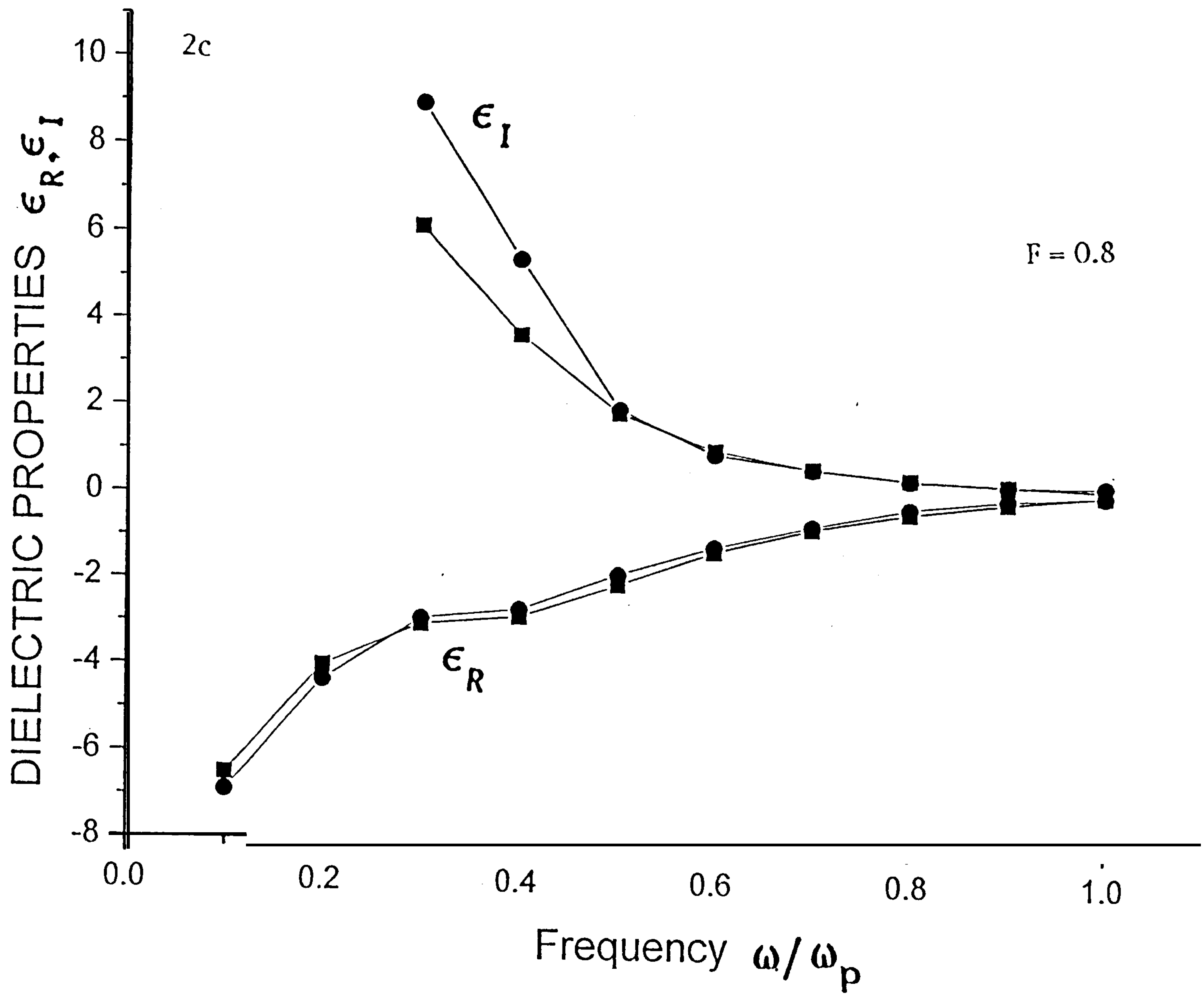


1c
ϵ_R
ϵ_I
F = 0.25
DIELECTRIC PROPERTIES ϵ_R, ϵ_I
Frequency, ω / ω_p
3.5
3.0
2.5
2.0
1.5
1.0
0.5
0.0
0.0
0.2
0.4
0.6
0.8
1.0



2a
ϵ_R
ϵ_I
F = 0.2
DIELECTRIC PROPERTIES ϵ_R, ϵ_I
8
6
4
2
0
-2
0.0
0.2
0.4
0.6
0.8
1.0
Frequency ω/ω_p


2b
DIELECTRIC PROPERTIES ϵ_R, ϵ_I
0
8
6
4
2
0
-2
0.0
0.2
0.4
0.6
0.8
1.0
Frequency, ω/ω_p
ϵ_I
ϵ_R
F = 0.5


2c
ϵ_I
ϵ_R
F = 0.8
DIELECTRIC PROPERTIES ϵ_R, ϵ_I
Frequency ω/ω_p
10
8
6
4
2
0
-2
-4
-6
-8
0.0
0.2
0.4
0.6
0.8
1.0